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Direct observations of the effects of aerosol loading on net ecosystem CO_2 exchanges over different landscapes

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[1] We present the first direct, multisite observations in support of the hypothesis that atmospheric aerosols affect the regional terrestrial carbon cycle. The daytime growing season (summer) CO_2 flux observations from six sites (forest, grasslands, and croplands) with collocated aerosol and surface radiation measurements were analyzed for high and low diffuse radiation; effect of cloud cover; and effect of high and low aerosol optical depths (AOD). Results indicate that, aerosols exert a significant impact on net CO_2 exchange, and their effect may be even more significant than that due to clouds. The response appears to be a general feature irrespective of the landscape and photosynthetic pathway. The CO_2 sink increased with aerosol loading for forest and crop lands, and decreased for grassland. The cause for the difference in response between vegetation types is hypothesized to be canopy architecture. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0315 Atmospheric Composition and Structure: Biosphere/atmosphere interactions; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 1610 Global Change: Atmosphere (0315, 0325); 1615 Global Change: Biogeochemical processes (4805). **Citation:** Niyogi, D., et al. (2004), Direct observations of the effects of aerosol loading on net ecosystem CO_2 exchanges over different landscapes, *Geophys. Res. Lett.*, *31*, L20506, doi:10.1029/2004GL020915.

1. Introduction

[2] Photosynthesis removes large amounts of CO_2 from the atmosphere. Net global terrestrial carbon exchange was nearly neutral in the 1980's, but resulted in a carbon sink in the 1990's [*Schimel et al.*, 2001]. CO_2 fertilization, land cover/land-use change, nitrogen loading, forest fires, and the regional hydrological cycle are some of the known factors affecting the carbon cycle [*Nemani et al.*, 2002]. Recent studies suggest that clouds and aerosols released in the atmosphere due to volcanic eruptions could also be important factors [*Gu et al.*, 2003; *Farquhar and Roderick*, 2003; *Krakauer and Randerson*, 2003].

[3] Given that previous studies cite significant events such as volcanic eruptions as the cause for variability in the carbon cycle, and that the mechanisms responsible for modified photosynthetic rates are modulated by aerosol loading, we ask the question: *can we detect the effect of relatively routine aerosol variability on field measurements of CO_2 fluxes, and if so, how does the variability in aerosol loading affect CO_2 fluxes over different landscapes*? Further, since studies such as *Krakauer and Randerson* [2003] question the positive effects of aerosols on the terrestrial carbon cycle; and modeling analysis of *Cohan et al.* [2002] indicated that the aerosol effects on CO_2 fluxes could depend on cloudiness, we seek to find: *whether or not the direct observations indicate an increase or a decrease in field scale CO_2 fluxes?* Thus, even though the effects of clouds on CO_2 fluxes are well documented [*Hollinger et al.*, 1998; *Gu et al.*, 2002], studies linking direct observations of aerosol loading on surface CO_2 fluxes are lacking. Using field measurements, we present additional evidence of the importance of aerosol feedback on regional climate via the biogeochemical pathways affecting the terrestrial carbon cycle.

2. Data and Methods

[4] We used CO_2 flux (*Fc*) data from the AmeriFlux network [*Baldocchi et al.*, 2001], and cloud-free aerosol optical depth (AOD) data from NASA Aerosol Robotic Network: AERONET [*Holben et al.*, 2001] for assessing the effect of aerosol loading on net ecosystem exchange (NEE). Six locations had concurrent *Fc* and AOD observations. The landscapes (locations and period with concurrent AOD and *Fc* data) were: broadleaf deciduous forest (Walker Branch, TN, 2000), mixed forest (Willow Creek/Lost Creek, WI, 2000–01), crops (winter wheat: Ponca, OK, 1998–99; alternate soybean or corn: Bondville, IL, 1998–2002), and grassland (Barrow, AK, 1999; Shidler, OK, 1998–99).

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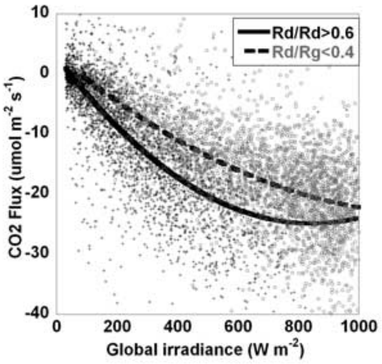

Figure 1a. Observed 30-minute averaged daytime observations of CO_2 flux and global irradiance for 1996–2000 summer (June–August). 2nd polynomial best fit (n = 3177, $p < 0.05$) are shown. Solid line: high diffuse regimes (Rd/Rg > 0.6, r = 0.78); dashed line: low diffuse radiation regime (Rd/Rg < 0.4, r = 0.67). See color version of this figure in the HTML.

[5] All data were quality assured by graphical and statistical means. Periods with either *Fc* or AOD measurements missing were eliminated. Daytime observations (since solar radiative effects will be studied) from June through August were selected. This period corresponds to the growing season, and includes the peak photosynthetic activity and capacity of the canopy.

[6] Data were clustered into three pairs to test the sensitivity of the *Fc* to the diffuse radiative flux fraction (DRF), and then to assess the impact of clouds as well as aerosol loading on the *Fc*.

[7] In analysis I, all observations, regardless of cloud cover or aerosol loading were clustered according to DRF [calculated as ratio of diffuse (Rd) to global irradiance (Rg)]. Data with Rd/Rg > 0.6 were labeled high diffuse regime, and those with Rd/Rg < 0.4 were labeled as a low diffuse regime. For analysis II, data were clustered according to cloudy and non-cloudy sky conditions. This was determined by analyzing the global irradiance time series plots, GOES visible cloud images, and weather reports for each day [*Gu et al.*, 1999]. In analysis III, observations were analyzed according to the AOD values, for clear sky (i.e., no clouds) conditions.

3. Results and Discussion

[8] Since similar analysis was conducted for each site, we will discuss the results for one site (Walker Branch) in detail and summarize the results for all the sites.

3.1. Effect of Diffuse Radiation

[9] Figure 1a shows the observed daytime *Fc* (30-min averages) for high and low DRF clusters (i.e., Analysis I) for the 1996 to 2000 summer (June–August). The *Fc* increase in magnitude (in the figures a negative value indicates a net flux into the vegetation, i.e., a sink) as a function of Rg. Additionally, for the same irradiance, the surface *Fc* increases (in magnitude) with increasing DRF. For example, for Rg = 500 Wm^{-2}, the *Fc* is 13 $\mu molm^{-2}s^{-1}$ for low DRF, and 20 $\mu molm^{-2}s^{-1}$ i.e., about 50% higher, for the high DRF. Observations for the three summer months over five years clearly indicate a significant increase in daytime NEE for larger DRF for similar global irradiance.

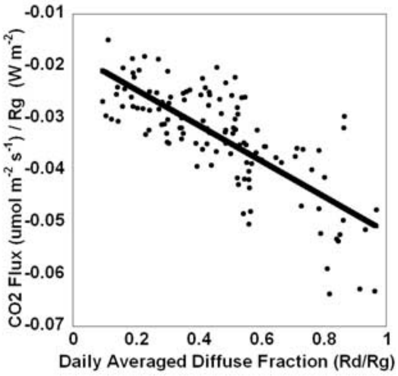

Figure 1b. Normalized daily CO_2 fluxes and diffuse fractions from the period shown in Figure 1a.

[10] The data from 1000 to 1600 LT, shown in Figure 1a were further averaged to yield a 'daily' value. The early morning and evening period were eliminated to avoid confounding due to low solar angles on high DRF caused independent of cloud cover or aerosol loading [*Gu et al.*, 1999]. The 'daily' averaged data are plotted in Figure 1b, with the *Fc* values normalized by Rg. A linear relation is obtained between higher DRF and the *Fc* values (both normalized for Rg). The best fit (n = 178, r = 0.75, $p < 0.05$) indicates that higher DRF enhances photosynthetic fluxes by about 30% at this study site.

3.2. Effect of Clouds

[11] The effect of increased mid-day DRF on *Fc* values (Figures 1a–1b) can be related to increased cloud cover and/or aerosol loading. Typically, for cloudy, overcast conditions the DRF is close to one. Hence the *Fc* data were clustered into 'clear' (i.e., non-cloudy) and 'cloudy' regimes (i.e., Analysis II, Figure 2a). The results are similar to those obtained in prior studies [*Hollinger et al.*, 1994; *Gu et al.*, 1999; *Roderick et al.*, 2001]. That is, under cloudy conditions the *Fc* values are larger for similar Rg. As in

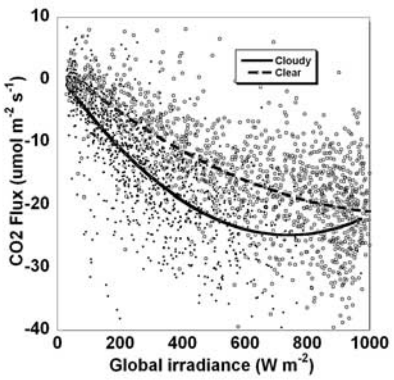

Figure 2a. Effect of cloudiness on CO_2 flux. Solid line: 'Cloudy' sky (r = 0.75, n = 1278); dashed line: 'Clear' (non-cloudy) sky (r = 0.74, n = 1395). See color version of this figure in the HTML.

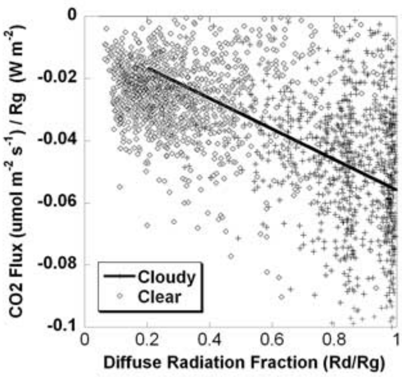

Figure 2b. Normalized 30-minute averaged CO_2 flux and diffuse radiation fraction (DRF) clustered for cloudy (+) and non-cloudy conditions. Solid line is best fit (r = 0.14, n = 2479). See color version of this figure in the HTML.

Analysis I, the 30-minute *Fc* values were normalized by Rg (Figure 2b). The normalized *Fc* show two distinct clusters for cloudy and clear (non-cloudy) sky conditions. CO_2 fluxes under cloudy conditions (i.e., high DRF) are typically larger than those under non-cloudy conditions with the same Rg (i.e., lower DRF).

3.3. Effect of Aerosol Loading

[12] As seen in Figures 2a–2b, some cloud-free days also have relatively high DRF values. This can be due to aerosols. Hence, the clear sky conditions data were analyzed further as a function of aerosol loading (through aerosol optical depth: AOD). For the Walker Branch site, coincident *Fc* and AOD data were available for June and July 2000 (Figure 3).

[13] The results indicate that the CO_2 flux is typically higher for larger aerosol loading. Another noteworthy feature of Figure 3 is that even under high aerosol loading, the Rg can be high ($\sim$900 Wm^{-2}).

[14] The increase in *Fc* with aerosol loading is likely to be the result of larger DRF. Consequently, the effect of AOD on DRF was studied. DRF shows a near linear relation with aerosol loading (r = 0.92, n = 119, not shown). Therefore, the variation in CO_2 fluxes can be considered to be an indirect

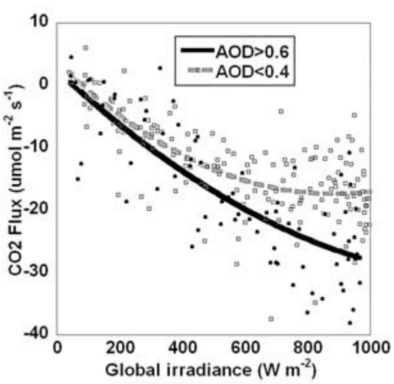

Figure 3. 30-minute averaged daytime observations of CO_2 flux and global irradiance during June–July of 2000. 2nd polynomial best fit is shown (n = 255), solid: AOD > 0.6 (r = 0.81); dashed: AOD < 0.4 (r = 0.72).

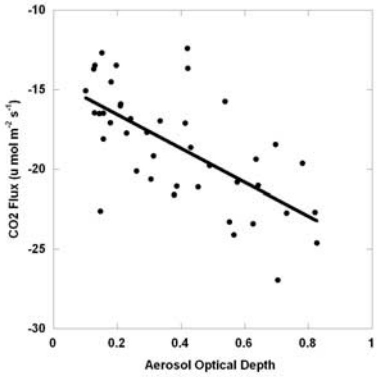

Figure 4. Aerosol optical depth (AOD) and CO_2 flux at the Walker Branch site for June–July 2000. With increasing aerosol loading, the landscape is a larger CO_2 sink. Solid line is best fit (n = 43, r = 0.76). See color version of this figure in the HTML.

effect of increased regional aerosol loading. Indeed, a nearly linear relation is obtained between AOD and the *Fc* under the clear sky conditions (Figure 4). With increasing AOD, the surface CO_2 fluxes are consistently larger.

3.4. Comparing the Effect of Aerosol Loading on CO_2 Fluxes Over Different Landscapes

[15] To investigate the regional effect of aerosol loading on field-scale NEE, the analysis described for the Walker Branch forest site was repeated for the five other sites (Figure 5). The sites represented the following landscapes (and photosynthetic pathways): winter wheat (C3), corn (C4), soybean (C3), grasslands (C3/C4), and mixed forest (C3).

[16] For all the sites, field-scale NEE varies with AOD, and every landscape has a different response. The woody (Figure 5a) and agricultural (Figure 5b) sites show an increase in the field- scale CO_2 flux 'sink' due to aerosol loading. Interestingly, both the grassland sites (Figure 5c) show an opposite response, and indicate a decreased field-scale CO_2 flux 'sink' with increased aerosol loading. Reviewing the best fits, the effect of aerosol loading on *Fc* is largest for C4 grassland (Shidler), and crops (corn; Bondville 1999, 2001), and relatively less on C3 crops (winter wheat, Ponca; and soybean, Bondville 1998, 2000, 2002). The *Fc* measurements over trees (forest sites) are also sensitive to the aerosol loading. Thus, both the canopy architecture (and hence the canopy scale radiative feedback on photosynthesis) and the photosynthesis pathway appear to be important factors. Additionally, there is significant scatter in the relationship between AOD and *Fc*, indicating other environmental variables (beyond aerosol loading) also influence the results. Indeed for all the landscapes, those variables that are known to affect photosynthesis rates (such as leaf area index and soil moisture availability) were also found to be significant in modulating the CO_2 fluxes (not shown). The results, however, clearly indicate that aerosol loading has a significant impact on the net ecosystem CO_2 exchange over terrestrial landscapes.

[17] For the results discussed above (Figure 5) 500 nm AOD data were chosen since it corresponded to a PAR wavelength. The AERONET AODs are centered over seven wavelengths (340 nm to 1020 nm), and the analysis was extended for all AOD wavelengths.

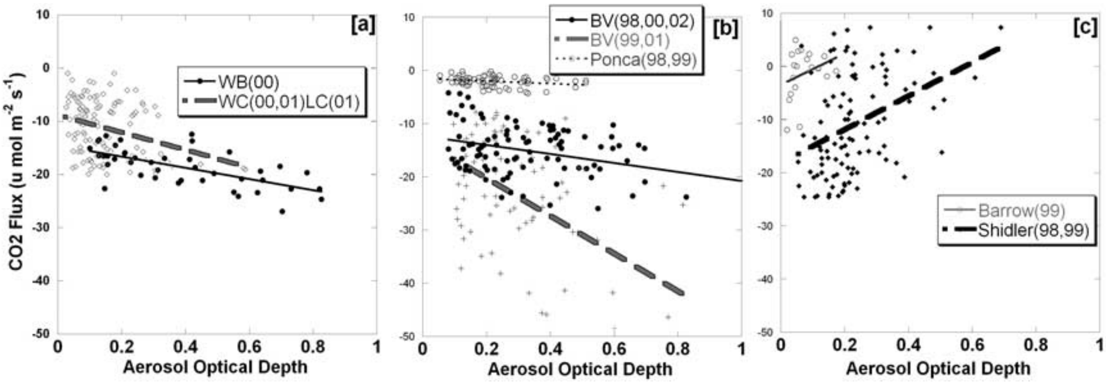

Figure 5. Relation between aerosol optical depth (AOD) and CO_2 flux at different sites and landscapes. The best fits and the periods for which data were used are also shown. (a) Trees- WB: Walker Branch (r = 0.76, n = 43); WC,LC: Willow Creek, Lost Creek (r = 0.52, n = 124); (b) Crops - BV: Bondville (r = 0.44, n = 150, for BV(98,00,02); n = 82, r = 0.61 for BV(99, 01); and (c) Grass- Barrow (r = 0.39, n = 23) and Shidler (r = 0.55, n = 124). Aerosols can increase (decrease) CO_2 fluxes/sink potential over forest and croplands (grasslands). C4 vegetation [Shidler; BV(99,01)] show largest sensitivity, C3 crops/grasslands show least, and trees show a moderately high influence of aerosols on CO_2 fluxes. See color version of this figure in the HTML.

[18] The AOD – CO_2 flux relation is sensitive to the choice of wavelength used and different landscapes may show sensitivity to different wavelengths. For example, the slope of the CO_2 flux – AOD best fit varies between −1.4 (for 340 nm) to −1.54 (for 1020 nm) radians for the deciduous forest site, and corresponding variation is comparatively less (from −1.53 to −1.55 radians) for the cropland. Thus NEE over a woody landscape could be even more sensitive to the aerosol loading than discussed in the analysis above. However, this does not alter the conclusion that aerosol loading can influence the terrestrial CO_2 fluxes.

4. Conclusions

[19] Study results suggest that aerosol induced radiative effect is an important modulator of regional carbon cycles. For the different study sites, DRF affected CO_2 fluxes, with an increase in DRF correlating with higher CO_2 flux values (sink) for trees and crops; and a lower sink for grasslands. The effect was clearly seen under cloudy conditions, during which the DRF was close to unity. It was also identified under higher aerosol loading in non-cloudy sky conditions. Aerosols can therefore routinely influence surface irradiance and hence the terrestrial CO_2 flux and regional carbon cycle.

[20] The reason for increased CO_2 fluxes with increasing DRF for forests and croplands is considered to result from an increase in the vegetative canopy fraction that is receiving illumination (without photosaturation). The advantage of increased diffuse radiation under clear sky, high aerosol conditions does not appear to be available to grasslands due to the canopy architecture. Additional confounding effects due to temperature, and humidity might exist and should be explored. Data from the early morning and evening period will also have high DRF but were eliminated in our analysis. For the whole-day carbon exchange, other factors that alter plant response may offset the effects of aerosol loading.

[21] Aerosols are abundant in the environment and their effects on climate are only poorly understood. Aerosols can alter irradiance at the top of atmosphere and even more profoundly at the surface and affect the biosphere [*Schwartz*, 1996]. Changes in DRF, due to aerosol loading, appear to have the potential to alter the terrestrial carbon exchange.

[22] Past studies on the impact of diffuse radiation on CO_2 flux focused on either the effect of cloudiness [e.g., *Hollinger et al.*, 1998] or the impact of volcanic aerosols [*Gu et al.*, 2003] on CO_2 flux exchange. *Gu et al.* [2002] analyzed field measurements across the United States and showed that NEE was larger for a higher diffuse fraction of the incoming radiation. Thus, even though field and model studies provide increasing evidence that photosynthesis rates and NEE will increase with DRF, the majority of these studies have been based on either episodic analysis of aerosol loading (i.e., effect of the Mt. Pinatubo eruption) or as the effect of cloudiness (which are conditions of high DRF). Thus our study is the first multi-site, observational analysis to investigate effects of persistent regional aerosol loading (which typically has a lifetime on the order of a week) on field NEE. Such an analysis is important for several reasons. First, the results provide evidence that routine aerosol loading due to natural or anthropogenic sources have the potential to influence regional CO_2 flux. Second, even though past studies indicate that for a given irradiance, NEE would be higher under cloudy conditions; cloudiness in itself may not be the dominant factor that increases the ability of a region to be a carbon sink. This is because even though it has a large DRF, total radiation is dramatically reduced. Hence cloudiness could even lead to lower, rather than higher, NEE over the region [cf. *Krakauer and Randerson*, 2003]. Alternatively, our results indicate that increasing aerosol loading will increase the diffuse fraction of the radiation without significantly reducing the total radiation itself and could be a prominent forcing affecting the CO_2 flux variability over a region. Thus, the potential of the vegetated land surface to be a sink for atmospheric carbon could depend on regional aerosol loading.

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